Study on the Electrodriven Action of Gelatin Hydrogel in Silicone Oil

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ABSTRACT: Action of gelatin hydrogel crosslinked with glutaraldehyde under dc electric field of high voltage was studied. The experimental results show that the hydrogel can be driven by dc electric field of high voltage in the medium of silicone oil, and the action of the hydrogel is composed of rotation and translation. There is a low critical applied voltage of 0.5 kV, below which the action of the hydrogel may not be observed. The motion velocity and angle of the rotation of gelatin hydrogel are influenced by the electric field intensity. Because silicone oil is very stable and hard to electrolyze in electric field, gas formation associated with the electrolysis of water, which is inevitable and disadvantageous in conventional system of electric-stimulus-response hydrogel, would not happen. Our work has provided a novel kind of electrodriven methods for hydrogels. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 100: 1664–1667, 2006

Key words: hydrogel; gelatin; electric field driving; rotation and translation; silicone oil

INTRODUCTION

Intelligent hydrogel is a novel kind of soft and wet material, which shows the ability of changing its volume, shape, and phase reversibly to external stimulus in environmental conditions, such as changes in pH, temperature, ionic strength, solvent composition, and electromagnetic radiation, etc.[1–8] Over the recent decades, intelligent hydrogel has attracted much attention in their potential application areas, such as flexible actuators, electromechanical engines, artificial muscles, sensors, and biomedical devices, etc.[9–13]

Among them, the electric-field-responsive hydrogel seems to be particularly interesting in connection with the fact that the electric field is the most conventional and convenient stimulus from the point of signal control. But, conventional systems of electric-stimulus-response hydrogel is in aqueous solution,[14–20] though the electrolysis of water plays an important role in the electric-stimulus-response behavior of hydrogel, gas generation from the electrode is a serious problem in practical application. To solve the problem of electrolysis, Osada[21] and Hiral[22] developed electroresponsive organogels made by swelling in organic polar solvents and had obtained significant results. The organic polar solvents used are dimethylformamide (DMF)[21] and dimethylsulfoxide (DMSO).[22] Although DMF and DMSO are hard to electrolyze and are relatively stable in electric field, they are affected with moisture easily. The presence of trace amount of water contained in the organic solvents and organogels would affect the electroresponsive behavior of gel badly, and so they must be dried before use and cannot be used in damp environment. Thus, this brings great inconvenience to their applications.

We have studied the electro-stimulative response of gelatin hydrogel crosslinked with glutaraldehyde in NaCl solution.[23] In this study, we have searched for a novel and completely different electrodriven method for gel actuation from the conventional method, and reported the action of gelatin hydrogel driven by dc electric field of high voltage in the medium of silicone oil.

EXPERIMENTAL

Materials

Gelatin (chemical grade), with an isoelectric point of about 4.9, was purchased from the Qingdao Chemicals (Qingdao, China). Dimethyl silicone oil (ε_f = 2.60

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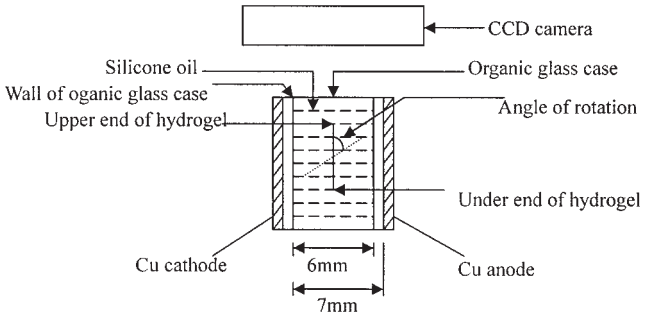

Figure 1 System used for measuring the motion of gelatin hydrogel.

–2.80, $\sigma_f = 10^{-12}$–10^{-13} s m^{-1}, $\rho = 0.965$–0.975 g cm^{-3}, $\eta = 500$ mPa s, 25°C) was purchased from Shanghai Chemicals (Shanghai, China). Glutaraldehyde aqueous solution (25%) is of analytical grade. All reagents were used without further purification.

Preparation of gelatin hydrogel

Gelatin (0.6 g) was dissolved in 6 mL of distilled water at 45°C. After dissolving completely, 1.0 mL of 1% glutaraldehyde aqueous solution was added to the gelatin solution under agitation. Then, glass capillaries with an internal diameter of 1.0 mm were immersed in the solution, where gelation occurred within 2 min. After 24 h, the gels were removed from the capillaries and cut into segments of 4 mm long in cylindrical form for the following experiment.

Action measurement of gelatin hydrogel under dc electric field in silicone oil

The apparatus used for measuring the motion of gelatin hydrogel under dc electric field is schematically shown in Figure 1. Dimethyl silicone oil was poured into a plastic case equipped with two parallel copper electrodes. The electrodes were fixed to the outside walls of the case. The walls of the case are 1 mm wide, and the distance between the two electrodes was 8 mm. The cylindrical gelatin hydrogel was placed in the center of the case and immersed in the silicone oil. The action of the gel was triggered by applying dc voltage up to 5.0 kV and monitored by a digital video system.

RESULTS AND DISCUSSION

The action of the hydrogel in silicone oil under electric stimulus is composed of rotation and translation (Fig. 2), and there is a low critical applied voltage of 0.5 kV, below which the action of the hydrogel may not be observed. Rotation and translation of the hydrogel will be discussed further.

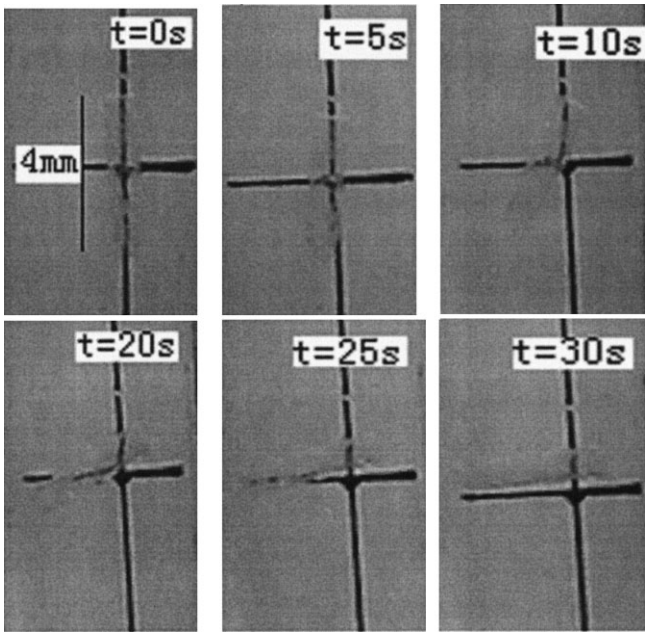

Figure 2 Photographs of gelatin hydrogel showing the rotation and translation under 2 kV.

Rotation of the hydrogel in electric field is investigated by recording its angle of rotation at time t under different applied voltages. Figure 3 shows the relationship between the angle of rotation as a function of time and the voltage of the applied electric field. As shown in Figure 3, the gradient slope in the plot of the angle of rotation versus time became steeper with increasing applied voltage, and then leveled off at a steady state. Figure 4 shows the curve of maximum angle of rotation versus applied voltage. The maximum angle of rotation increases with the applied voltage up to 2 kV. When it is more than 2 kV, the maximum angle of rotation is kept constant (= 90°). It meant that when the axes of hydrogel paralleled to the direction of electric field, the rotation of hydrogel would not con-

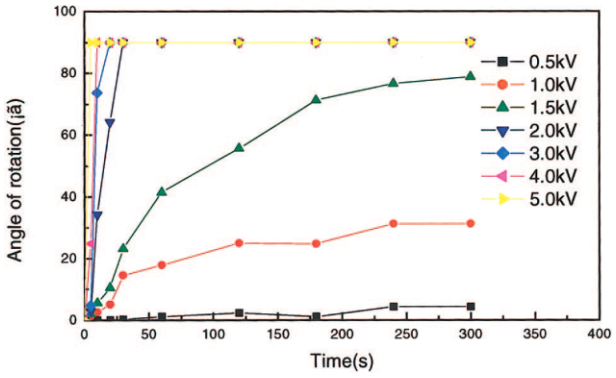

Figure 3 Angle of rotation of gelatin hydrogel versus time under different voltages. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

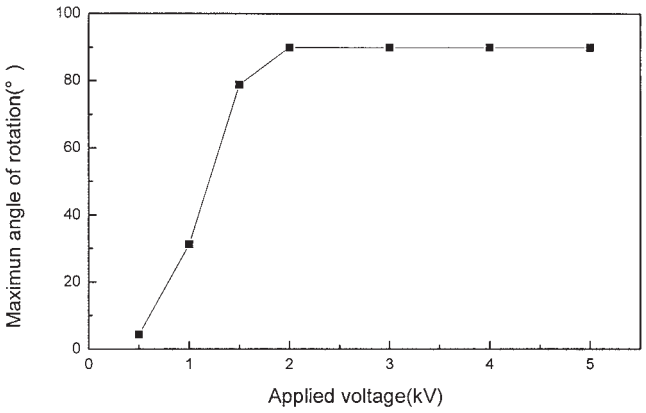

Figure 4 Effect of the applied voltage on the maximum angle of rotation of gelatin hydrogel.

tinue even if the applied voltage continued to be increased.

Figure 5 shows the relationship between the applied voltage and the time when gelatin hydrogel reached its maximum angle of rotation. The speed of rotation increased with the increase of applied voltage, but the speed increased more slowly when the applied voltage is more than 2 kV. It only took 4.5 s for the hydrogel to reach its maximum angle of rotation of 90° under 5.0 kV.

The two ends of the hydrogel were named as upper and under end, respectively. Translation of hydrogel in electric field is investigated by recording its two ends' displacements at time t, under different applied voltages. The original position of the ends is defined as coordinate origin. The sign of the displacement is expressed positive when the end moves to the right of its original position. On the contrary, the sign of the displacement is expressed negative when the end moves to the left of its original position. Figure 6 shows the displacement of the two ends of the hydrogel versus time under different applied voltage. When the applied voltage is not more than 1 kV, translation of the whole hydrogel occurred, but its rotation is inconspicuous. When the applied voltage is higher than 1.5 kV, the rotation became conspicuous and more symmetrical, and both speeds of translation and rotation increased with the increase in applied voltage.

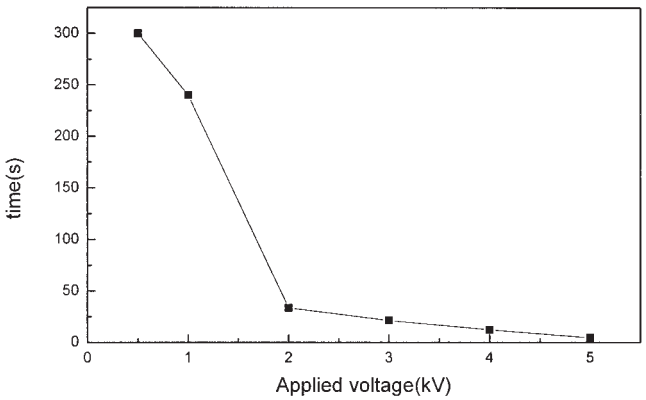

Figure 5 Relationship between the applied voltage and the time when gelatin hydrogel reached its maximum angle of rotation.

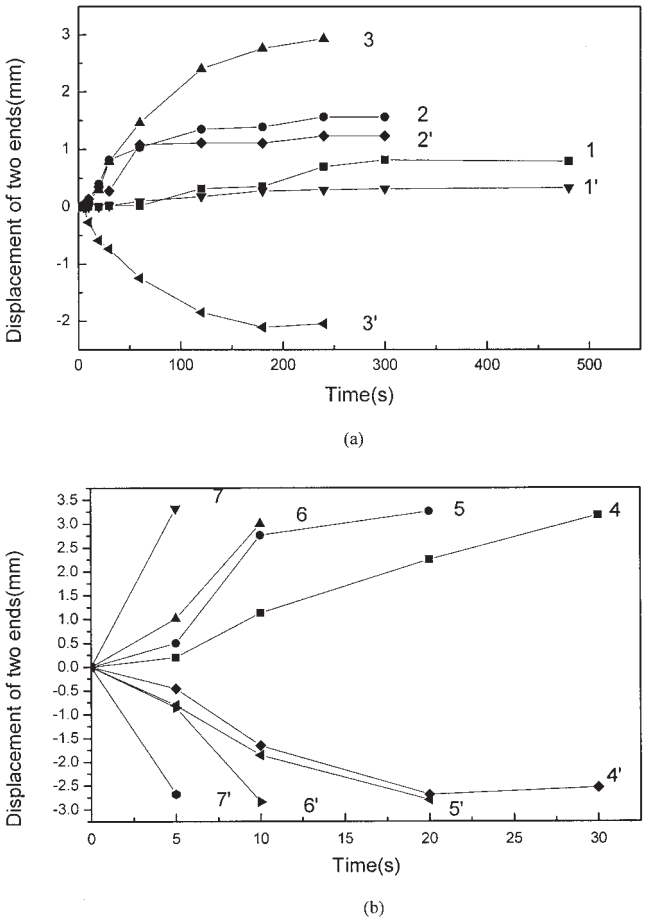

Figure 6 Displacement of the two ends of the hydrogel versus time under different applied voltage. Upper end: (1) 0.5 kV, (2) 1 kV, (3) 1.5 kV, (4) 2 kV, (5) 3 kV, (6) 4 kV, (7) 5 kV; Under end: (1′) 0.5 kV, (2′) 1 kV, (3′) 1.5 kV, (4′) 2 kV, (5′) 3 kV, (6′) 4 kV, (7′) 5 kV.

As we mentioned earlier, when 0.5 kV $\leq V \leq$ 1.5 kV, the maximum angles of rotation are all below 90°, and the motion of the hydrogel, including translation and rotation, would stop when the maximum angle of rotation is reached. But, when $V \geq$ 2.0 kV, the axis of the hydrogel turned 90° vertically until it paralleled to the direction of electric field. The rotation of the hydrogel stopped, while its translation continued by means of moving to and fro between the two electrodes at that time. The frequency of moving enhanced with the increase of applied voltage. The time was cut off as soon as the hydrogel reached its maximum angle of rotation of 90° [Fig. 6(b)].

In addition, the reversibility of the movement of the hydrogel under electric field was examined. When the axis of the hydrogel was placed not parallel to the electrodes before applying electric field, the hydrogel always turned to the direction that would minimize its moment of force after being subjected to electric field. If the polarity of the electric field was altered, the direction of movement including rotation and translation would not change. This means that the movement of the hydrogel was irreversible. If the applied voltage was higher than 2.0 kV, it would turn until the angle of rotation reached 90°, and the moment of force becomes 0. We also observed similar action when the silicone oil was replaced by transformer oil, under dc electric field. But, if the silicone oil in this case was replaced by NaCl aqueous solution, according to our experimental method, the hydrogel would remain motionless in NaCl solution because the electrodes did not contact with the NaCl aqueous solution and no electrochemical reactions occurred on the electrodes. Gelatin hydrogel is hydrophilic and it cannot be swollen in hydrophobic silicone oil. Apparently, the mechanism of action of hydrogel in nonaqueous medium is different from that of conventional electroresponsive hydrogel completely, and cannot be explained by Flory's theory of the osmotic pressure.[16,24,25]

According to electrical phenomena at interfaces,[26] there is a electric double layer at the interfaces between the gelatin hydrogel and silicone oil. The primary mechanism of action of the hydrogel in silicone oil may be based on a distortion of the double layer when subjected to electric field. Because the formation of double layer is universal, the action of the hydrogel in nonaqueous medium under electric field should be universal too. In fact, we did observe the similar action of different materials (such as poly(vinyl alcohol) hydrogel, wire of copper, sheets of organic glass, paper, and wood, etc.) in silicone oil under dc electric field of high voltage. The only difference among their action is the difference in speeds of rotation and translation according to different materials. Further study, which is under progress in our laboratory, is necessary for the clarification of the mechanism of the action.

CONCLUSIONS

Gelatin hydrogel crosslinked with glutaraldehyde can be driven by dc electric field of high voltage in the medium of silicone oil, and the action of the hydrogel is composed of rotation and translation. There is a low critical applied voltage of 0.5 kV, below which the action of the hydrogel may not be observed. The motion velocity and angle of the rotation of gelatin hydrogel are influenced by the electric field intensity. Because silicone oil is very stable and hard to be affected with damp and electrolyzed in electric field, electrolysis of water and gas formation is avoided. Our work has provided a novel kind of electrodriven methods for hydrogels. Because many different kinds of materials all show the similar action, this discovery may provide adequate materials to choose for designing micromachine and flexible actuator, etc.

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